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(54) Title: METHOD OF FORMING SINGLE CRYSTALS OF A CERAMIC, SEMICONDUCTIVE OR MAGNETIC MATERIAL

(57) Abstract: The invention is concerned with a method of forming a single crystal of a ceramic, semiconductive or magnetic material. The method according to the invention comprises the steps of (a) compacting a nanocrystalline powder comprising particles having an average particle size of 0.05 to 20 µm and each formed of an agglomerate of grains with each grain comprising a nanocrystal of a ceramic, semiconductive or magnetic material; and (b) sintering the compacted powder obtained in step (a) at a temperature sufficient to cause an exaggerated growth of at least one of the grains, thereby obtaining at least one single crystal of aforesaid material. Instead of sintering the compacted powder, it is also possible to contact same with a template crystal of the aforesaid material, and to heat the compacted powder and template crystal in contact with one another so as to cause a sustained directional growth of the template crystal into the compacted powder, thereby obtaining a single crystal having a size larger than the template crystal. By using nanocrystalline powders, the temperature of operation for crystal growth is reduced, the rate of crystal growth increases, and crystals with large size and with very little or no porosity or inclusions can be obtained.

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METHOD OF FORMING SINGLE CRYSTALS OF A CERAMIC, SEMICONDUCTIVE OR MAGNETIC MATERIAL

TECHNICAL FIELD

The present invention pertains to improvements in the field of
5 single crystals. More particularly, the invention relates to an improved
method of forming single crystals of a ceramic, semiconductive or magnetic
material.

BACKGROUND ART

Large size single crystals are of great interest in electronic
10 and optical applications. Single crystals are produced using different
techniques such as top-seeded solution growth (TSSG), templated grain
growth (TGG) and exaggerated grain growth (EGG). Due to difficulties
inherent to these fabrication methods, the commercial cost of single crystals
is relatively high.

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The TSSG technique involves bringing a seed which is a
single crystal into contact with a melt of the material having the same
composition as the single crystal to be produced. The seed is brought
slowly into contact with the surface of the melt, then it is rotated and pulled
20 up. Since the temperature of the seed is lower than that of the melt, the
atoms of the melt join the surface of the seed and crystallize on the seed. By
turning and pulling the seed, the latter grows and forms a solid droplet. The
bottom of this droplet is always in contact with the melt. The problems
encountered in TSSG include:

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1. High operating temperature: the starting material must melt and this causes serious problems when the melting point is too high.
2. Strict temperature control: crystal growth occurs within a narrow range of temperature. If the temperature is higher than this range, the seed melts and the contact between the seed and the melt is cut. If the temperature is lower than this range, a sudden undesirable growth occurs and it is possible that the solid be full of solution inclusions, voids and polycrystalline material.
3. Strict control of cooling and pulling rates: pulling and cooling rates are very sensitive to the solid droplet diameter. Moreover, during radial expansion, it is possible that solution trapping or incomplete crystalline formation may occur. These malformed facet intersections can be avoided by gradually decreasing the cooling rate; however, this requires strict control of cooling rate and long run duration.
4. Lack of diameter control and the formation of a solution droplet on the bottom of the solid droplet, which may cause cracking.

The TGG technique involves contacting a template crystal and a sintered polycrystalline matrix and then heating the template crystal and polycrystalline matrix in contact with one another to produce a single crystal via sustained directional growth of the template crystal into the polycrystalline matrix. The driving force for boundary migration is provided by the grain boundary free energy of the polycrystalline matrix. The problems encountered in TGG include:

1. Boundary migration rates and, consequently, template growth are relatively slow because the matrix consists of grains with large size (micron size) which reduces considerably the driving force for template growth.

- 5 2. Low driving force and long diffusion paths contribute to increase the temperature necessary for TGG. In general, grain growth occurs within the polycrystalline matrix itself during TGG and reduces the template growth rate considerably.

10 The EGG technique involves essentially the sintering of a polycrystalline powder at a temperature sufficient to cause some grains to grow abnormally to much large size than the average due an enhanced material transfer in some directions and on some specific planes. Admixing additives can help the exaggerated grain growth. For example, addition of a
15 small amount of SiO_2 or TiO_2 enhances the exaggerated grain growth of BaTiO_3 . It has also been reported that placing several seeds (single crystals with a size larger than the powder particle size) in the powder before sintering enhances the exaggerated growth of the seeds. The problems encountered in EGG include:

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1. There is no shape control of the final crystal.
2. Since the starting powder contains large particles (micron size), the diffusion rate is slow and this reduces considerably the driving force for
25 crystal growth. Consequently, the rate of crystal growth is too small.

3. A small amount of porosity is present in the grains due to pore trapping within the crystal. Elimination of these pores is very difficult (sometimes impossible) because of the long diffusion paths.
- 5 4. The maximum size of single crystal produced by this method is relatively small. The growth rate is high in the early stages of sintering, but it reduces very rapidly by a further increase in particle size.

DISCLOSURE OF INVENTION

It is therefore an object of the invention to overcome the
10 above drawbacks and to provide an improved method of forming single crystals of a ceramic, semiconductive or magnetic material.

According to one aspect of the invention, there is provided a method of forming a single crystal of a ceramic, semiconductive or
15 magnetic material, in accordance with the EGG technique. Such a method comprises the steps of:

a) compacting a nanocrystalline powder comprising particles having an average particle size of 0.05 to 20 μm and each formed of an
20 agglomerate of grains with each grain comprising a nanocrystal of a ceramic, semiconductive or magnetic material; and

b) sintering the compacted powder obtained in step (a) at a temperature sufficient to cause an exaggerated growth of at least one of the
25 grains, thereby obtaining at least one single crystal of the aforesaid material.

According to another aspect of the invention, there is provided a method of forming a single crystal of a ceramic, semiconductive or magnetic material, in accordance with the TGG technique. Such a method comprises the steps of:

5

a) compacting a nanocrystalline powder comprising particles having an average particle size of 0.05 to 20 μm and each formed of an agglomerate of grains with each grain comprising a nanocrystal of a ceramic, semiconductive or magnetic material;

10

b) contacting the compacted powder obtained in step (a) with a template crystal of the aforesaid material; and

c) heating the compacted powder and template crystal in contact with one another to cause a sustained directional growth of the template crystal into the compacted powder, thereby obtaining a single crystal having a size larger than the template crystal.

The term "nanocrystal" as used herein refers to a crystal having a size of 100 nanometers or less.

Nanocrystalline powders exhibit good sinterability. They can be prepared by different techniques such as those described for example in US Patent Nos. 5,514,349 and 5,958,348. They can also be prepared by a technique called "high-energy ball milling", as described in Applicant's Canadian Patent Application No. 2,331,470 filed on January 19, 2001 and corresponding to International Application No. PCT/CA02/00070. Depending on the type of the material and the technique of production, the particle size of nanocrystalline powders may lie in the

range of 0.05 to 20 μm . When the particles are nanometric in size, the specific area of the powder in this case is very high (20-400 m^2/g). However, when the particles are larger, they contain several nanosized crystallites. In such a case, although the specific area of powder is not very
5 high, the material consists of very large quantity of grain boundaries.

Having a large surface area or large quantity of grain boundaries enhances the diffusion rate. In addition, high quantity of grain boundaries, with higher free energy, compared to the grain itself, increases
10 the driving force for densification and grain growth during sintering.

Another factor influencing the driving force for densification and grain growth is the surface energy. Small nanosized grains having a small curvature radius are unstable at high temperatures and possess high
15 chemical potentials. So they have a tendency to join on the flat surfaces or those with large curvature radii in order to minimize the overall free energy.

For all the above reasons, the crystal growth from nanocrystalline powders is rapid and takes place at lower temperatures. By
20 using nanocrystalline powders, the temperature of operation for crystal growth is reduced, the rate of crystal growth increases, and crystals with large size and with very little or no porosity or inclusions can be obtained.

MODES FOR CARRYING OUT THE INVENTION

Examples of ceramic materials from which single crystals
25 may be formed include aluminum oxide, aluminum nitride and silicon nitride. On the other hand, examples of semiconductive material include zinc oxide and compounds having the formula $\text{Ba}_x\text{Ti}_y\text{O}_z$ wherein x and y

each range from 0.1 to 20 and z ranges from 0.3 to 60, such as BaTiO_2 and $\text{Ba}_3\text{Ti}_4\text{O}_{11}$. Where the semiconductive material is a compound of the formula $\text{Ba}_x\text{Ti}_y\text{O}_z$, the nanocrystalline powder of such a material can be obtained by subjecting barium oxide and titanium dioxide to high-energy ball milling to cause solid state reaction therebetween and formation of particles having an average particle of 0.05 to 20 μm , each particle being formed of an agglomerate of grains with each grain comprising a nanocrystal of a compound of the formula $\text{Ba}_x\text{Ti}_y\text{O}_z$. In the particular case of barium titanate (BaTiO_3), the nanocrystalline powder can be obtained by subjecting a barium titanate powder having an average grain size larger than 1 μm to high-energy ball milling to cause formation of particles having an average particle size of 0.05 to 20 μm , each particle being formed of an agglomerate of grains with each grain comprising a nanocrystal of barium titanate.

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Examples of magnetic materials include compounds having the formula $\text{Sm}_2\text{Fe}_x\text{Co}_{17-x}\text{N}_y$ wherein $0 \leq x \leq 17$ and $0 \leq y \leq 3$, such as $\text{Sm}_2\text{Fe}_{17}$, $\text{Sm}_2\text{Fe}_{17}\text{N}_3$, $\text{Sm}_2\text{Co}_{17}$ and $\text{Sm}_2\text{Co}_{17}\text{N}_3$. It is also possible to use a compound of the formula $\text{Nd}_2\text{Fe}_x\text{B}_y$ wherein $9 < x < 19$ and $0.3 < y < 3$, such as $\text{Nd}_2\text{Fe}_{14}\text{B}$.

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The expression "high-energy ball milling" as used herein refers to a ball milling process capable of forming the aforesaid particles comprising nanocrystalline grains of the ceramic, semiconductive or magnetic material, within a period of time of about 40 hours.

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Where the EGG technique is followed, a grain growth enhancing agent or a seed crystal of the ceramic, semiconductive or

magnetic material is preferably added to the nanocrystalline powder, prior to step (a). For example, silica or titanium dioxide can be added in an amount of 0.01 to 8 wt.% to enhance the exaggerated grain growth of BaTiO₃. Step (b), on the other hand, is preferably carried out at a
5 temperature ranging from 0.5 T_m to 0.95 T_m, where T_m is the melting point of the ceramic, semiconductive or magnetic material.

The method of the invention also allows producing very homogeneously doped single crystals. Sometimes, single crystals are doped
10 with elements, ions or compounds in order to modify the optical and electrical properties. In some cases, the doping elements may have a concentration gradient within the single crystal. The use of nanocrystalline powders allows one to prepare very homogeneous powder where the doping elements are distributed in nanometer scale. Growing a single crystal from
15 such a homogenous powder results in a crystal having a very high homogeneous concentration of doping element.

The following non-limiting examples illustrate the invention.

20 **EXAMPLE 1**

A coarse-grained BaTiO₃ powder (99.9% pure) having an average grain size larger than 1 μm was used as starting material. 10 g of this BaTiO₃ powder were milled in a steel crucible using a SPEX 8000
25 (trademark) vibratory ball mill operated at 16 Hz. After 10 hours of high-energy ball milling, a nanocrystalline BaTiO₃ powder having a particle size between 1 and 5 μm and a mean crystallite size smaller than 100 nm was obtained. The nanocrystalline powder was then pressed uniaxially at a

pressure of 250 MPa using a cylindrical die having 1 cm in diameter. The compacted powder thus obtained was sintered at a temperature of 1300°C for a period of 6 hours. A heating rate of 5°C/min. was used. A polycrystalline bulk material was obtained. A few grains grew to a large size (several millimeters).

EXAMPLE 2

A coarse-grained BaTiO₃ powder (99.9% pure) having an average grain size larger than 1 µm was used as starting material. 3.96 g of this BaTiO₃ powder and 0.04 g of stearic acid were milled in a silicon nitride crucible using a SPEX 8000 (trademark) vibratory ball mill operated at 16Hz. After 10 hours of high-energy ball milling, a nanocrystalline BaTiO₃ powder having a mean crystallite size smaller than 100 nm was obtained. The nanocrystalline powder was then uniaxially pressed at a pressure of 250 MPa using a cylindrical die having 1 cm in diameter. The compacted powder thus obtained was sintered at a temperature of 1130° C for a period of 10 hours. A heating rate of 5° C/min was used. A polycrystalline bulk material was obtained. A few grains grew to a large size (several millimeters).

EXAMPLE 3

20

A BaTiO₃ single crystal was prepared according to the same procedure as described in Example 1 or 2 and under the same operating conditions, with the exception that 0.02 g of silica were admixed with the coarse-grained powder, prior to compaction.

25

EXAMPLE 4

A BaTiO₃ single crystal was prepared according to the same procedure as described in Example 1 or 2 and under the same operating conditions, with the exception that a seed crystal of BaTiO₃ having a mean diameter of about 1 μm was placed in the coarse-grained powder, prior to compaction.

EXAMPLE 5

10

A BaTiO₃ single crystal was prepared according to the same procedure as described in Example 1 or 2 and under the same operating conditions, with the exception that prior to compaction, 0.02 g of titanium dioxide were admixed with the coarse-grained powder and a seed crystal of BaTiO₃ having a mean diameter of about 1 μm was then placed in the powder.

EXAMPLE 6

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A nanocrystalline BaTiO₃ powder was produced by ball milling 7.26 g of BaO and 2.397 g of TiO₂ in a steel crucible using a SPEX 8000 vibratory ball mill operated 16 Hz. After 10 hours of high-energy ball milling, a nanocrystalline powder consisting of BaTiO₃ and having a particle size varying between 1 and 5 μm was obtained. The crystallite size, measured by X-ray diffraction, was about 20 nm. The nanocrystalline powder was then pressed uniaxially at a pressure of 250 MPa using a cylindrical die having 1 cm in diameter. The compacted powder thus obtained was sintered at a temperature of 1300°C for a period of 6 hours. A

heating rate of 5°C/min. was used. A polycrystalline bulk material was obtained. A few grains grew to a large size (several millimeters).

EXAMPLE 7

5 A nanocrystalline $\text{Ba}_3\text{Ti}_4\text{O}_{11}$ powder was produced by ball milling 7.26 g of BaO and 3.196 g of TiO_2 in a steel crucible using a SPEX 8000 vibratory ball mill operated 16 Hz. After 10 hours of high-energy ball milling, a nanocrystalline powder consisting of $\text{Ba}_3\text{Ti}_4\text{O}_{11}$ and having a particle size varying between 1 and 5 μm was obtained. The crystallite size, measured by X-ray diffraction, was about 20 nm. The nanocrystalline powder was then pressed uniaxially at a pressure of 250 MPa using a cylindrical die having 1 cm in diameter. The compacted powder thus obtained was sintered at a temperature of 1300°C for a period of 6 hours. A heating rate of 5°C/min. was used. A polycrystalline bulk material was
15 obtained. A few grains grew to a large size (several millimeters).

EXAMPLE 8

 A thin film of BaTiO_3 was deposited on a MgO substrate by
20 chemical deposition to form a template crystal of BaTiO_3 . A nanocrystalline BaTiO_3 powder produced by high-energy ball milling as described in Example 1 or 6 was pressed uniaxially at a pressure of 250 MPa using a cylindrical die having 1 cm in diameter. The compacted powder thus obtained was placed on the BaTiO_3 thin film and the combination was
25 heated at a temperature of 1200°C to cause a sustained directional growth of the template crystal in the compacted powder. A single crystal of BaTiO_3 having a size larger than the template crystal was obtained.

EXAMPLE 9

The surface of a BaTiO₃ single crystal prepared in accordance with any one of Examples 1 to 6 were polished. The single crystal was placed at the center of a die and the void in the die around the crystal was filled with nanocrystalline BaTiO₃ powder containing a dopant element in a predetermined concentration. The powder was then pressed isostatically at a pressure of 250 MPa. The compacted powder was sintered at 1300°C for a period of 6 hours. These steps were repeated with different concentrations of dopant element in order to obtain several layers of dopant having a concentration gradient around the single crystal.

EXAMPLE 10

A thin film of BaTiO₃ was deposited on a MgO substrate by chemical deposition to form a template crystal of BaTiO₃. A nanocrystalline powder produced by high-energy ball milling as described in Example 2 was pressed axially at a pressure of 250 MPa using a cylindrical die having 1 cm in diameter. The compacted powder thus obtained was placed on the BaTiO₃ thin film and the combination was heated at a temperature of 1130°C to cause a sustained directional growth of the template crystal in the compacted powder. A single crystal of BaTiO₃ having a size larger than the template crystal was obtained.

CLAIMS

1. A method of forming a single crystal of a ceramic, semiconductive or magnetic material, comprising the steps of:
 - 5 a) compacting a nanocrystalline powder comprising particles having an average particle size of 0.05 to 20 μm and each formed of an agglomerate of grains with each grain comprising a nanocrystal of a ceramic, semiconductive or magnetic material; and
 - 10 b) sintering the compacted powder obtained in step (a) at a temperature sufficient to cause an exaggerated growth of at least one of said grains, thereby obtaining at least one single crystal of said material.
2. A method according to claim 1, wherein prior to step (a), a grain growth enhancing agent is added to said nanocrystalline powder.
- 15 3. A method according to claim 1, wherein prior to step (a), a seed crystal of said material is added to said nanocrystalline powder.
4. A method according to claim 1, wherein said ceramic, semiconductive or magnetic material has a melting point and wherein step
20 (b) is carried out at a temperature ranging from $0.5 T_m$ to $0.95 T_m$, where T_m is the melting point of said material.
5. A method according to claim 1, wherein each said grain
25 comprises a nanocrystal of a ceramic material.

6. A method as claimed in claim 5, wherein said ceramic material is selected from the group consisting of aluminum oxide, aluminum nitride and silicon nitride.
- 5 7. A method according to claim 1, wherein each said grain comprises a nanocrystal of a semiconductive material.
8. A method according to claim 7, wherein said semiconductive material is barium titanate or zinc oxide.
- 10 9. A method according to claim 7, wherein said semiconductive material is barium titanate and wherein, prior to step (a), a grain growth enhancing agent is added to said nanocrystalline powder.
- 15 10. A method according to claim 9, wherein said grain growth enhancing agent comprises silica or titanium dioxide.
11. A method according to claim 7, wherein said semiconductive material is barium titanate and wherein said nanocrystalline powder is
20 obtained by subjecting a barium titanate powder having an average grain size larger than 1 μm to high-energy ball milling to cause formation of particles having an average particle size of 0.05 to 20 μm , each particle being formed of an agglomerate of grains with each grain comprising a nanocrystal of barium titanate.
- 25 12. A method according to claim 7, wherein said semiconductive material is a compound of formula $\text{Ba}_x\text{Ti}_y\text{O}_z$ in which x and y each range from 0.1 to 20 and z ranges from 0.3 to 60, and wherein said

nanocrystalline powder is obtained by subjecting barium oxide and titanium dioxide to high-energy ball milling to cause solid state reaction therebetween and formation of particles having an average particle of 0.05 to 20 μm , each particle being formed of an agglomerate of grains with each
5 grain comprising a nanocrystal of a compound of the formula $\text{Ba}_x\text{Ti}_y\text{O}_z$.

13. A method according to claim 12, wherein said semiconductive material is $\text{Ba}_3\text{Ti}_4\text{O}_{11}$.

10 14. A method according to claim 1, wherein each said grain comprises a nanocrystal of a magnetic material.

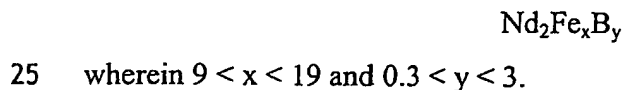
15. A method according to claim 14, wherein said magnetic material is a compound of the formula:



wherein $0 \leq x \leq 17$ and $0 \leq y \leq 3$.

16. A method according to claim 15, wherein said magnetic material is a compound selected from the group consisting of $\text{Sm}_2\text{Fe}_{17}$,
20 $\text{Sm}_2\text{Fe}_{17}\text{N}_3$, $\text{Sm}_2\text{Co}_{17}$ and $\text{Sm}_2\text{Co}_{17}\text{N}_3$.

17. A method according to claim 14, wherein said magnetic material is a compound of the formula:



18. A method according to claim 17, wherein said magnetic material is $\text{Nd}_2\text{Fe}_{14}\text{B}$.

19. A method according to claim 1, wherein said nanocrystalline powder has an average particle size ranging from 1 to 5 μm .
- 5 20. A method of forming a single crystal of a ceramic, semiconductive or magnetic material, comprising the steps of:
- a) compacting a nanocrystalline powder comprising particles having an average particle size of 0.05 to 20 μm and each formed of an agglomerate of grains with each grain comprising a nanocrystal of a ceramic, semiconductive or magnetic material; and
 - 10 b) contacting the compacted powder obtained in step (a) with a template crystal of said material; and
 - c) heating the compacted powder and template crystal in contact with one another to cause a sustained directional growth of the template crystal into the compacted powder, thereby obtaining a single crystal
 - 15 having a size larger than said template crystal.
21. A method according to claim 20, wherein each said grain comprises a nanocrystal of a ceramic material.
- 20 22. A method as claimed in claim 21, wherein said ceramic material is selected from the group consisting of aluminum oxide, aluminum nitride and silicon nitride.
- 25 23. A method according to claim 20, wherein each said grain comprises a nanocrystal of a semiconductive material.

24. A method according to claim 23, wherein said semiconductive material is barium titanate or zinc oxide.

25. A method according to claim 23, wherein said
5 semiconductive material is barium titanate and wherein said nanocrystalline powder is obtained by subjecting a barium titanate powder having an average grain size larger than $1\mu\text{m}$ to high-energy ball milling to cause formation of particles having an average particle size of 0.05 to $20\mu\text{m}$, each particle being formed of an agglomerate of grains with each grain
10 comprising a nanocrystal of barium titanate.

26. A method according to claim 23, wherein said semiconductive material is a compound of formula $\text{Ba}_x\text{Ti}_y\text{O}_z$ in which x and y each range from 0.1 to 20 and z ranges from 0.3 to 60, and wherein said
15 nanocrystalline powder is obtained by subjecting barium oxide and titanium dioxide to high-energy ball milling to cause solid state reaction therebetween and formation of particles having an average particle of 0.05 to $20\mu\text{m}$, each particle being formed of an agglomerate of grains with each grain comprising a nanocrystal of a compound of the formula $\text{Ba}_x\text{Ti}_y\text{O}_z$.

20

27. A method according to claim 26, wherein said semiconductive material is $\text{Ba}_3\text{Ti}_4\text{O}_{11}$.

28. A method according to claim 20, wherein each said grain
25 comprises a nanocrystal of a magnetic material.

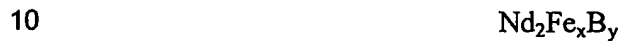
29. A method according to claim 28, wherein said magnetic material is a compound of the formula:



wherein $0 \leq x \leq 17$ and $0 \leq y \leq 3$.

30. A method according to claim 29, wherein said magnetic
5 material is a compound selected from the group consisting of $\text{Sm}_2\text{Fe}_{17}$,
 $\text{Sm}_2\text{Fe}_{17}\text{N}_3$, $\text{Sm}_2\text{Co}_{17}$ and $\text{Sm}_2\text{Co}_{17}\text{N}_3$.

31. A method according to claim 28, wherein said magnetic
material is a compound of the formula:



wherein $9 < x < 19$ and $0.3 < y < 3$.

32. A method according to claim 31, wherein said magnetic
material is $\text{Nd}_2\text{Fe}_{14}\text{B}$.

- 15 33. A method according to claim 20, wherein said nanocrystalline
powder has an average particle size ranging from 1 to 5 μm .

INTERNATIONAL SEARCH REPORT

International Application No

PCT/CA 02/00168

A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 C30B1/12 C30B29/32 C30B29/52 C30B29/38

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC 7 C30B

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	YOO Y-S ET AL: "Fabrication of BaTiO3 Single Crystals by Using the Exaggerated Grain Growth Method" JOURNAL OF THE EUROPEAN CERAMIC SOCIETY, ELSEVIER SCIENCE PUBLISHERS, BARKING, ESSEX, GB, vol. 17, no. 14, 1997, pages 1725-1727, XP004097468 ISSN: 0955-2219 the whole document --- -/--	1,2,5, 7-12

☒ Further documents are listed in the continuation of box C.☒ Patent family members are listed in annex.

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C.(Continuation) DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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